Electron Transport in Thin Insulating Films*

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Abstract—Experiments on the electron transport through thin insulating barriers have been performed with diodes of Ta-Ta_2,O_5-Au, Ta-Ta_2O_5-Al, Ta-Ta_2O_5-Bi, Zn-ZnO-Au, Al-Al_2O_3-Al, and Al-Al_2O_3-Au. The analysis of the dependence of current on temperature and film thickness allows a distinction of two cases: imperfect and perfect dielectrics. In the former case, the mechanism for electron transport is field ionization of trap-type states at low temperatures, and thermal ionization of this state at higher temperatures. In the latter case, Schottky-emission and field-emission have been observed.

When a large electric field is applied between two metal electrodes separated by a thin insulating film, a current I is observed to flow, which increases in a generally exponential way with the applied voltage V. This current can often be described to a reasonable degree of accuracy over several decades by the expression $I \approx \exp$ (Const. $V^{\frac{1}{2}}$). The purpose of this paper is to review in a general way some of the processes which can give rise to such a current, and to discuss a few of the interesting basic physical questions related to such currents.

The first comment which must be made is that there are a very large number of phenomena which can give rise to currents of this general form. The first question one must answer in any specific situation is whether the current is limited by the rate at which current can flow through the bulk of the dielectric itself, or by the rate at which carriers can be injected from the electrode(s) into the dielectric. Examples of bulk-limited operation are the various regimes of space-charge-limited current; examples of electrode-limited operation are Schottky emission and Fowler-Nordheim tunneling. Although this distinction is easily stated, it is often difficult to determine experimentally. To illustrate the problems arising in bulk-limited operation, we will discuss the case of Ta_2O_5, an insulator commonly employed as a capacitor dielectric. The I–V characteristic of a typical Ta-Ta_2O_5-Au sample is shown in Fig. 1. The linearity of the Log I vs. $V^{\frac{1}{2}}$ plot might, in the absence of other information, lead one to believe that the current is due to electrode-limited Schottky emission. However, the currents persist with the same dependence upon electric field up to thicknesses where space-charge limitations must be important. Hence, one suspects that even at the lowest thicknesses, the current is bulk-controlled. In such a case, the first thing to establish is whether the barriers at the two interfaces are different. The abrupt drop in current at approximately $\frac{1}{2}$ Volt indicates a built-in contact potential of this amount, consistent with the difference between the Ta and Au work functions. A check of the effect is made by noting the dependence of this offset voltage on the metal electrode. In this case, Ta-Ta_2O_5-Au gave the offset shown, Ta-Ta_2O_5-Al gave zero offset voltage. When it is established that there is a built-in contact potential in the sample, the next step is to measure the voltage required for a given current as a function of insulator thickness for both polarities of applied bias. The results of this procedure are shown in Fig. 2. In the case of an electrode-limited process, the line would not have unity slope, while with the bulk-limited process a straight line is expected with unity slope

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and intercept twice the contact potential, which is the observed result. The detailed mechanism of the bulk limitation needs further elaboration. The general equations of space-charge-limited operation cannot be solved in closed form, but approximate solutions can be obtained under various limiting conditions. If one assumes a density of traps in the insulator much greater than the number of trapped carriers which, in turn, is much greater than the density of free carriers, and assumes that the carriers are liberated from the traps by a field-emission process at low temperatures and by thermal emission at high temperature, one obtains a space-charge potential maximum very close to one electrode followed by a region of very nearly-constant field extending to the other electrode. This result is consistent with the experimental results. It should be noted that this interpretation would indicate that the author's measurement of the anomalous intercept in the capacitance and I–V data on very thin Ta-Ta_2O_5-Au samples should be interpreted in terms of this space-charge maximum, rather than in terms of electric-field penetration of the electrodes, as was thought previously. The large effect found in Ta-Ta_2O_5-Bi may still be related to the field-penetration problem.

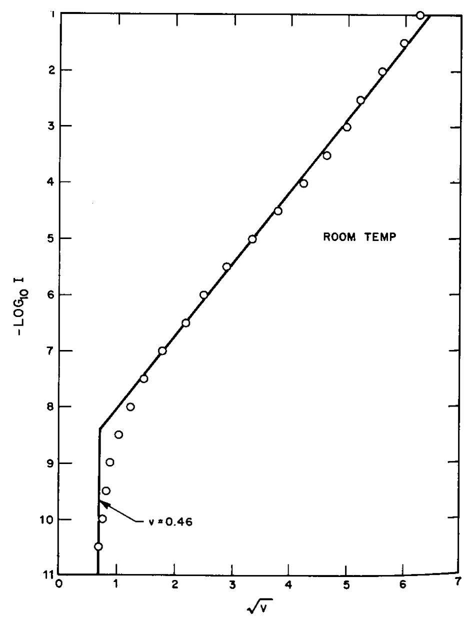

Figure 1: I–V characteristic of Ta-Ta_2O_5-Au sample (from [1]).

Although these results are not new, they have been reviewed here in order to emphasize the degree of caution necessary in the interpretation of data on these dielectric films.

A more fundamental and interesting question is the current expected from a dielectric film sufficiently perfect and sufficiently thin that ordinary space-charge type bulk limitations are not important. Under these conditions, the current can be generally classified as due to either thermionic or field emission. This is clearly not a strict distinction since there is the same broad crossover region as in metal-vacuum emission.

Although there have been numerous reports of currents following the Schottky law, to the author's knowledge, a clear demonstration of the Schottky effect in thin film structures has not previously been published.

For this reason, I am including some new data taken at our laboratory which we believe indicates true Schottky emission. The image force lowering $\Delta\varphi$ of the potential barrier φ depends upon the electric field

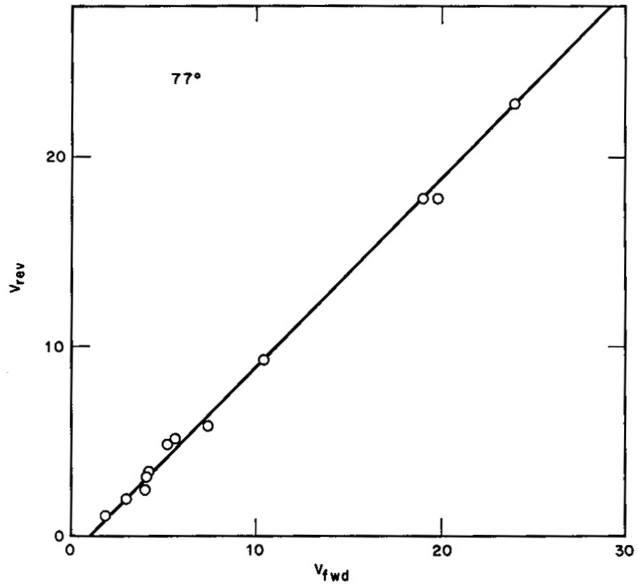

Figure 2: Voltage of each polarity required for a given current in samples of different thickness (from [1]).

F as $\Delta\varphi = \left(\frac{qF}{4\pi\epsilon}\right)^{\frac{1}{2}}$. This lowering gives rise to a voltage-current characteristic of the form

$$\mathrm{J} = \mathrm{AT}^2 \exp\left(\frac{\mathrm{q}\varphi}{\mathrm{kt}}\right) \exp\left(\frac{\mathrm{qF}}{4\pi\epsilon}\right)^{\frac{1}{2}}. \tag{1}$$

Thus, a plot of current vs. the square root of the voltage should yield a straight line over many decades. If the dielectric constant of the material is known, one can infer a thickness from the slope of this curve by relating the field to the voltage. One can also determine the thickness from the capacitance of the sample. These measurements should agree within experimental error. Fig. 3 shows some data on a Zn-ZnO-Au sample taken at room temperature. The straight line corresponds to a thickness of 61 $\mathring{A}$, whereas, the thickness as measured by capacitance was 57 $\mathring{A}$, indicating agreement within the errors in measuring the sample area. In itself, this is good evidence that Schottky emission is involved. However, for a complete study one should compare the thermal activation energy with the barrier height as measured by photoemission, and also as inferred from the intercept of the Schottky plot. On the sample shown, the thermal activation energy gave approximately 0.75 eV, while the intercept indicated approximately 0.8 eV, depending slightly upon the effective mass in the ZnO. Although we cannot yet report acceptable photoemission measurements of barrier height on the thin film samples of this material, measurements on single crystal ZnO give barriers of the same general magnitude. Hence, one can conclude that Schottky emission has been unambiguously observed in these film structures as the dominant current flow mechanism at room temperature.

We will now focus our attention on field emission in thin film dielectrics, a topic of fundamental interest in which considerable good work has been done in many laboratories. Fig. 4 shows the energy band scheme for a typical metal-insulator-metal structure under small applied bias. We are considering sufficiently small temperatures (on sufficiently high barriers) that no appreciable current flows by thermal excitation over the barrier. Electron transfer between the metal electrodes can occur by electron tunneling through the "forbidden" gap of the insulator. By Bloch's Theorem, the wave function ψ of an electron in a crystal (considering one dimension only) can be written $\psi = \exp\,(\mathrm{ikx})\ \mathrm{u(x)}$ where k is the wave number of the electron, and u(x) is a function periodic in x with the period of lattice. In the "forbidden" gap, k is, in general, complex and the wave function is exponentially damped, as $\exp\,(-\alpha \mathrm{x})$ where α is the imaginary

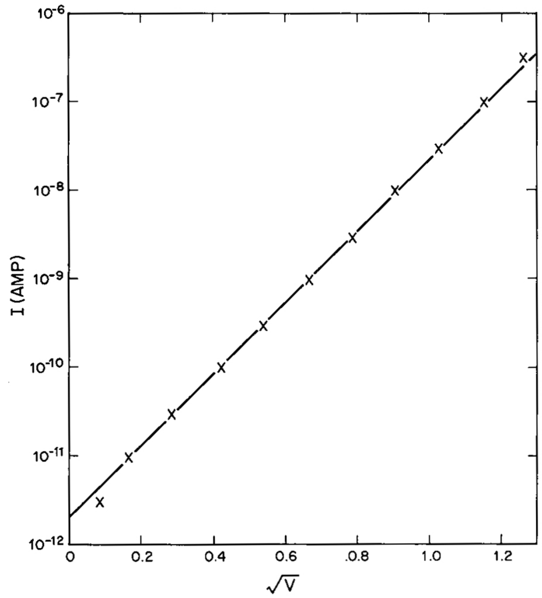

Figure 3: I–V curve of Zn-ZnO-Au sample at 300°K. The slope agrees very well with the theoretical value from Schottky emission.

part of the wave number. The probability per unit time that an electron at a given initial energy will make a transition from one electrode to the other will be proportional to (probability of electron in 1st electrode at that energy) $\cdot$ probability of an empty state in 2nd electrode at that energy) $\cdot$ exp $(-\int_0^t \alpha\, dx)$ where t is the thickness of the insulator. In order to calculate the total current, one must integrate this expression over all energies. In general, this program cannot be carried out in closed form, even if the dependence of α on energy and the two barrier energies are known. In the usual analysis of experimental results, the two barrier heights (and sometimes the thickness as well) are treated as adjustable parameters, while the dependence of α on energy is approximated by a parabolic relationship $E = h^2\alpha^2/2m^*$ (where the energy E is measured downward from the conduction band). Of course, m^* is not known independently for any of the materials such as Al_2O_3, and hence it too must be treated as an adjustable constant. With this variety of unknown parameters available for curve fitting, it is quite possible to fit I–V data over a limited range, even if the detailed nature of the phenomenon involved is not the same as the model used.

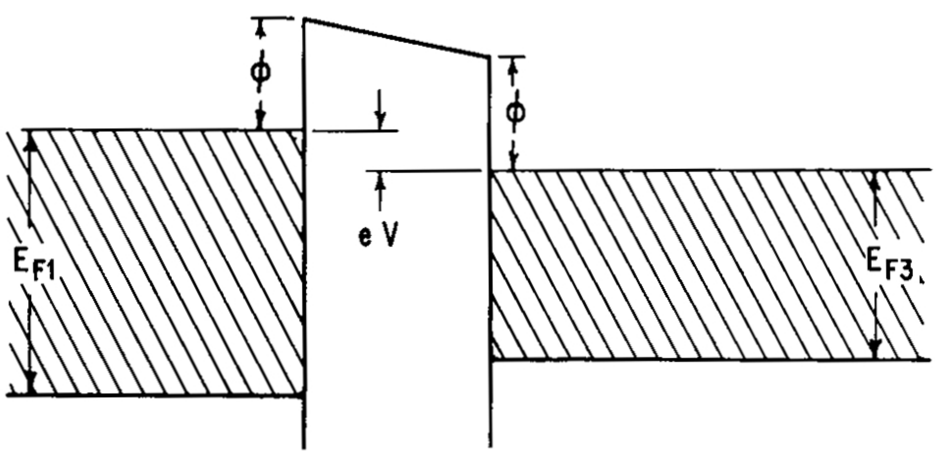

Figure 4: Energy band scheme for metal-dielectric-metal structure under small applied bias.

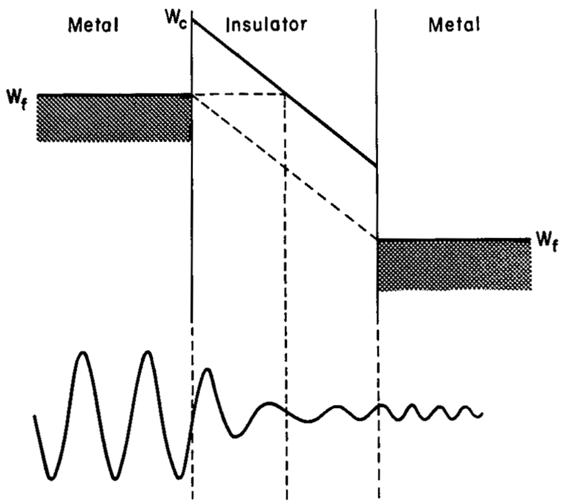

Figure 5: Energy band scheme for metal-dielectric-metal structure under large applied bias. Shown also is schematic representation of electron wave function.

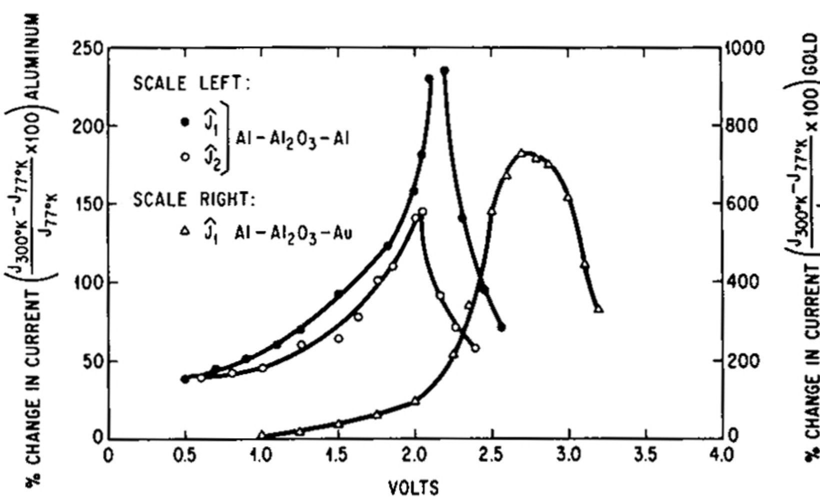

Figure 6: Temperature coefficient of tunnel current as a function of bias (from Ref. [2]).

The analysis must also include the image-force rounding of the barriers, an effect which becomes quite large at small thicknesses.

At this point, it should be clear that an increased understanding of the tunneling must be based upon careful experimental determination of the unknown parameters by means independent of the curve-fitting procedure. Table I shows the parameters involved and the independent measurements used in their determination. The barrier heights enter the I–V characteristic at low voltages only in that they are related to α through the effective mass, if such an approximation is valid. At higher voltages (Fowler-Nordheim region, see Fig. 5), the tunneling is directly into the conduction band and the barrier height determines the tunneling distance, and hence an internal consistency check is possible. When the applied voltage is equal to the 2nd barrier height, the temperature dependence exhibits a pronounced maximum (Fig. 6). This effect is independent of the detailed nature of the energy dependence upon α.

Table 1

Parameter	**Measurement**
Barrier Heights	Photoemission
	Cusps in Temp Dependence
α	Thickness Dependence
$\frac{\partial\,\alpha}{\partial\,\mathrm{E}}$	Temp Dependence

In addition, one can obtain a completely independent measure of the barrier heights using photoemission of electrons from the electrodes over the barrier into the insulator conduction band. If the structure is biased so as to collect electrons emitted from one electrode only, higher photon energies result in more electron injection. If the square root of the photocurrent is plotted vs. the photon energy (Fig. 7), a straight line results which can be extrapolated to zero photocurrent giving the barrier height. These measurements provide a value which can be compared directly with that obtained from the high-field I–V characteristic.

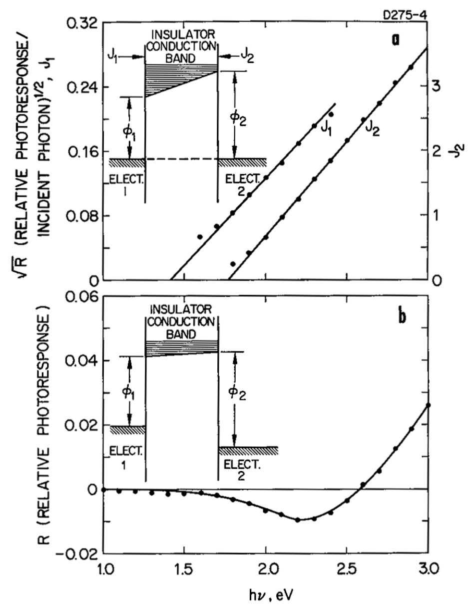

Figure 7: Photoemission determination of barrier heights on Al-Al_2O_3-Al tunnel structure (from Ref. [3]).

One of the most important parameters involved in the tunneling characteristic is the attenuation constant α. Curve-fitting procedures give very little information about this quantity unless its functional dependence upon energy is known. It is therefore very desirable to have at least a consistency check, and preferably an independent measurement of α as a function of energy. Although a completely independent evaluation of α is not possible, the variation of the I–V characteristic with thickness can be used to extract, nearly unambiguously, the dependence of α on energy. As an example of how this might be done, we write the tunneling probability $P = \exp\left(-\int_0^t \alpha \mathrm{dx}\right)$. If we have applied enough voltage so that the tunneling can be considered to be localized in a narrow energy range below the Fermi level of the first electrode, the total current can be written, approximately

$$J \propto \exp\left(-2\int_0^t \alpha\,\mathrm{dx}\right) \tag{2}$$

since α is a function of energy, we can change the variable of the integration

$$J \approx J_0 \exp\left(-2\mathrm{t}\int_{\varphi_1}^{\varphi_2-\mathrm{V}} \alpha\,(\mathrm{E})\mathrm{dE}\right) \tag{3}$$

or derivative with respect to voltage,

$$\frac{\mathrm{d}}{\mathrm{dV}}(-\frac{1}{2\mathrm{t}}\ \mathrm{Log}\ \frac{\mathrm{J}}{\mathrm{J}_0^0})_{\varphi_2-\mathrm{V}} = \alpha\,(\varphi_2 - \mathrm{V}) \tag{4}$$

This crude example illustrates how α as a function of energy can be extracted from the slope of the Log J vs. t-curves at different voltages.

One further cross-check is valuable to establish the consistency of the data in any given case. The temperature-dependence of the current at a given (low) voltage is due nearly completely to the change of α with energy. Thus, one is able to check this measurement against α(E) as determined by the thickness dependence. Consistency of all these measurements allows one a high degree of confidence that the model employed is indeed applicable to the experimental situation. Such a program requires a complete set of I–V curves as a function of both thickness and temperature, as well as photo-emission data, preferably as a function of voltage. However, it is the author's opinion that nothing short of such a program will clearly establish the full nature of the physical processes involved in thin film tunneling structures. We are just completing a study of this type on AlN films at the California Institute of Technology, and the results of this study will be published shortly.

References

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